LARGE-SCALE MERCURY CONTROL TECHNOLOGY TESTING FOR LIGNITE-FIRED UTILITIES – OXIDATION SYSTEMS FOR WET FGD

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ABSTRACT

The Energy & Environmental Research Center (EERC) is conducting a consortium-based effort directed toward resolving the mercury control issues facing the lignite industry. Specifically, the EERC team—the EERC, EPRI, URS, ADA-ES, Babcock & Wilcox, the North Dakota Industrial Commission, SaskPower, and the Mercury Task Force, which includes Basin Electric Power Cooperative, Otter Tail Power Company, Great River Energy, Texas Utilities (TXU), Montana—Dakota Utilities Co., Minnkota Power Cooperative, BNI Coal Ltd., Dakota Westmoreland Corporation, and the North American Coal Corporation—has undertaken a project to significantly and cost-effectively oxidize elemental mercury in lignite combustion gases, followed by capture in a wet scrubber. The applicability of this approach is expected to increase because of an expected demand for scrubbed systems in lignite utilities as well as subbituminous utilities in the United States and Canada. The oxidation process is proven at the pilot-scale and in short-term full-scale tests. Additional optimization is continuing on oxidation technologies, and this project focuses on monthlong full-scale testing.

The lignite industry has been proactive in advancing the understanding of and identifying control options for Hg in lignite combustion flue gases. Approximately 2 years ago, the EERC and EPRI began a series of Hg-related discussions with the Mercury Task Force as well as utilities firing Texas and Saskatchewan lignites. This project is one of three being undertaken by the consortium to perform large-scale Hg control technology testing to address the specific needs and challenges to be met in controlling Hg from lignite-fired power plants.

This project involves Hg oxidation upstream of a system equipped with an electrostatic precipitator (ESP) followed by wet flue gas desulfurization (FGD). The team involved in conducting the technical aspects of the project includes the EERC, Babcock & Wilcox, EPRI, URS, and ADA-ES. The host sites include Minnkota Power Cooperative Milton R. Young Station Unit 2 and TXU Monticello Unit 3. The work involves establishing Hg oxidation levels upstream of air pollution control devices (APCDs) and removal rates across existing ESP and FGD units, determining costs associated with those removal rates, investigating the possibility of the APCD acting as a multipollutant control device, quantifying the balance-of-plant impacts of the control technologies, and facilitating technology commercialization.

In the previous reporting period, a meeting on-site was conducted with MRY plant personnel to discuss the draft of the site specific test plan. The test plan was made available to plant personnel and project sponsors for their review and comments. In the current reporting period, these comments were incorporated into a final version of the test plan. Detailed planning for the testing phase of the Milton R. Young unit is well under way; it is scheduled to commence the last week in February 2005. In addition, corrosion probes were installed at the Milton R. Young Plant November 18, 2004. These will be removed in early January 2005 after 8 weeks of test exposure under baseline conditions.

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LIST OF ABBREVIATIONS

APCD air pollution control device

B&W Babcock & Wilcox BOP balance of plant

CMM continuous mercury monitor

COR contracting officer's representative

DOE U.S. Department of Energy

EERC Energy & Environmental Research Center EPA U.S. Environmental Protection Agency

ESP electrostatic precipitator

FF fabric filter

FGD flue gas desulfurization

ICR Information Collection Request (EPA)

MRY Milton R. Young Station

NDIC North Dakota Industrial Commission
NETL National Energy Technology Laboratory

OH Ontario Hydro
QA quality assurance
QC quality control
ROM run of mine

SCA specific collection area SDA spray dryer absorbers

SCEM semicontinuous emission monitor SEA sorbent enhancement additive

SOA solid oxidizing additive

TXU Texas Utilities

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EXECUTIVE SUMMARY

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1.0 INTRODUCTION

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The other projects cover sorbent injection technologies for systems equipped with ESPs and those equipped with spray dryer absorbers combined with fabric filters (SDA–FF) and an alternative oxidation technology. The overall intent of the proposed testing is to help maintain the viability of lignite-fired energy production by providing utilities with lower-cost options for meeting future Hg regulations.

2.0 BACKGROUND

Mercury is an immediate concern for the U.S. electric power industry because of the U.S. Environmental Protection Agency (EPA) December 2000 decision that regulation of Hg from coal-fired electric utility steam-generating units is appropriate and necessary under Section 112 of the Clean Air Act (1). EPA determined that Hg emissions from power plants pose significant hazards to public health and must be reduced. The EPA *Mercury Study Report to Congress* (2) and the *Utility Hazardous Air Pollutant Report to Congress* (3) identified coal-fired boilers as the largest single source of atmospheric Hg emissions in the United States, accounting for about one-third of the total anthropogenic emissions. On December 15, 2003, EPA published the proposed Utility Mercury Reductions Rule in order to solicit comments on multiple approaches for mercury emission control. EPA is currently reviewing comments on the proposed rule and is scheduled to put forth regulations in March 2005.

Even though Hg regulations for coal-fired utilities are imminent, significant issues remain and need to be resolved. The U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL) has acknowledged that data gaps exist for Hg control technologies for the immense U.S. reserves of lignite and subbituminous coals. The primary challenge is that these coals produce flue gases where difficult-to-control Hg⁰ is the dominant form. The EPA information collection request (ICR) indicates questions still exist regarding the impact of various APCDs and technologies for lignite-fired units on their ability to control Hg⁰ emissions. The lignite-based consortium believes there is a critical need for large-scale Hg oxidation testing at lignite-fired power plants equipped with an ESP and wet FGD. This project has been developed based on the input of consortium members and DOE guidance to address these issues.

In general, lignitic coals are unique because of highly variable ash content, ash that is rich in alkali and alkaline-earth elements, high oxygen levels, high moisture levels, and low chlorine content. Lignite coals typically contain comparable levels of Hg but significantly lower levels of chlorine compared to bituminous coals. Lignites have chlorine concentrations well below 200 ppm in the coal, whereas Appalachian and Illinois Basin bituminous coals can have chlorine levels in excess of 1000 ppm. These differences in composition have been shown to have important effects on the form of Hg emitted from a boiler and the capabilities of different control technologies to remove Hg from flue gas. Coals containing chlorine levels greater than 200 ppm typically produce flue gas dominated by more easily removable mercuric compounds (Hg²⁺), most likely mercuric chloride (HgCl₂). Conversely, experimental results indicate that lowchlorine (<50 ppm) coal combustion flue gases (typical of lignite) contain predominantly Hg⁰, which is substantially more difficult to remove than Hg^{2+} (3). Additionally, the generally high alkali and alkaline-earth contents of lignite coals may reduce the oxidizing effect of the alreadylow chlorine content by reactively scavenging chlorine species (Cl, HCl, and Cl₂) from the combustion flue gas. The level of chlorine in flue gases of recently tested lignites from North Dakota and Saskatchewan ranged from 2.6 to 3.4 ppmy, with chlorine contents ranging from 11 to 18 ppmw in the coal on a dry basis, respectively.

Few published data exist demonstrating the effectiveness of oxidation technologies for plants firing lignite coal. Lignite-fired power plants have shown a limited ability to control Hg emissions in currently installed ESPs, SDAs, and wet FGD systems (4). This low level of control

can be attributed to the high proportions of Hg^0 present in the flue gas. Typically, the form of Hg in the pulverized and cyclone-fired units is dominated by the Hg^0 content being greater than 85% of the total, and the average emitted from North Dakota lignite-fired power plants is roughly 6.3 lb/TBtu (4, 5). Figure 1 shows resulting Hg emissions measured using the Ontario Hydro (OH) method and continuous mercury monitors (CMMs) at the furnace exit during pilot tests at the EERC with North Dakota lignite. These results are consistent with the ICR results discussed above and with the recent baseline data for the proposed test sites, as shown later.

Hg oxidation technologies being investigated for lignites include catalysts and chemical agents. The catalysts that have been tested include selective catalytic reduction catalysts for NO_x reduction, noble metal-impregnated catalysts, and oxide-impregnated catalysts. The chemical agents include chlorine-containing salts (chloride compounds) and cofiring fuels that contain oxidizing agents (6, 7).

Theoretically, the use of chloride compounds to oxidize Hg^0 to Hg^{2^+} makes sense. The evidence includes chemical kinetic modeling of bench-scale test results, indicating that the introduction of chloride compounds into the high-temperature furnace region will likely result in the production of atomic chlorine and/or molecular chlorine, which are generally thought to be the dominant Hg^0 reactants in coal combustion flue gases (6). The formation of atomic chlorine is a key pathway involved in the chemical reaction mechanisms that result in the oxidation of Hg^0 (6). The pathway for Hg oxidation is gas-phase Hg^0 oxidation by atomic chlorine (chlorine radical). Recent kinetic modeling of chlorine radical formation as a function of temperature

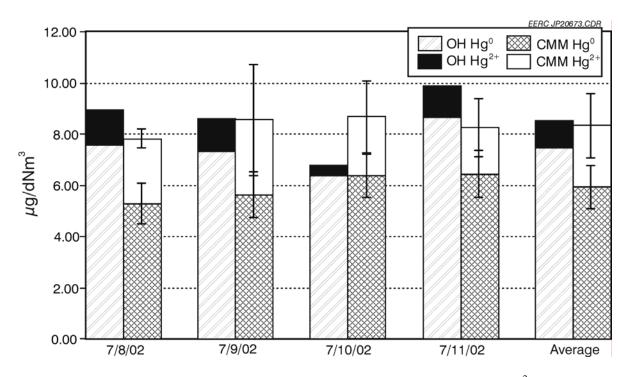


Figure 1. Inlet mercury speciation for Freedom, North Dakota, lignite ($\mu g/dNm^3 = microgram$ per dry normal cubic meter [corrected to 0°C and 3% O_2]).

and residence time is shown in Figure 2. The results indicate the importance of temperature in the abundance of chlorine radicals. Recent work, supported by EPRI, indicated that injection of HCl in lower-temperature regions downstream of the boiler was ineffective in oxidizing Hg^0 while injection of salt into the furnace resulted in significant oxidation (8).

Fuel additives for Hg oxidation have recently been tested in a pilot-scale system. Chemical additives or oxidants such as chloride salts have shown the ability to convert Hg⁰ to more reactive oxidized forms, as shown in Figure 3. In addition, recent EPRI short-term testing conducted at a 70-MW_e pulverized-coal-fired North Dakota power plant indicated the injection of chloride salts can result in increased Hg oxidation in the flue gas (8). Hg oxidation of up to 70% was observed at a salt injection rate that resulted in an HCl concentration of 110 ppm in the flue gas, as shown in Figure 4. In addition, the injection of salt resulted in enhanced removal of Hg across the SDA–FF, with removal efficiencies of up to 50% in short-term field testing (8).

Because of the promise seen in oxidation of Hg in flue gases produced from lignite coals, the project team is conducting long-term field testing of Hg oxidation and removal using a wet FGD at the Minnkota Power Cooperative MRY Unit 2 near Center, North Dakota, and the TXU Monticello Unit 3 near Mt. Pleasant, Texas.

MRY Unit 2 is a B&W Carolina-type radiant boiler designed to burn high-moisture, high-slagging/fouling North Dakota lignite. Nominally rated at 3,050,000 lb/hr, this unit is a cyclone-fired, balanced-draft, pump-assisted circulation boiler. The unit began commercial operation in May 1977 and is base-loaded at 450 MW gross. The unit is equipped with a cold-side ESP for particulate control and a wet FGD unit for SO₂ control. The cold-side ESP has a specific

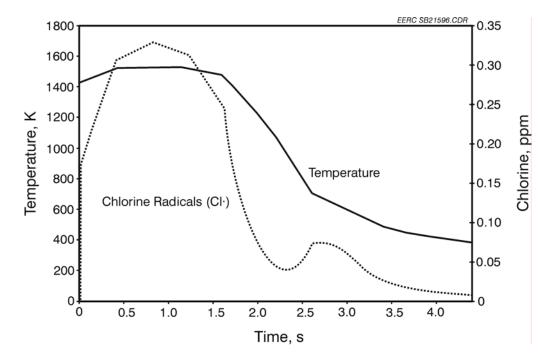


Figure 2. Prediction of chlorine radical formation as a function of temperature and residence time typical of a utility boiler using a kinetic mode (Chemkin).

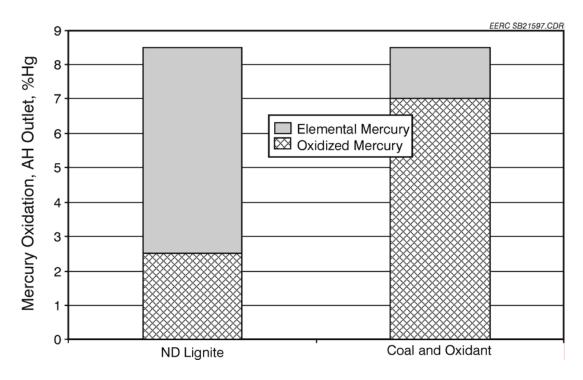


Figure 3. Oxidation of mercury through the addition of a chlorine-containing additive to the coal in EERC pilot-scale testing.

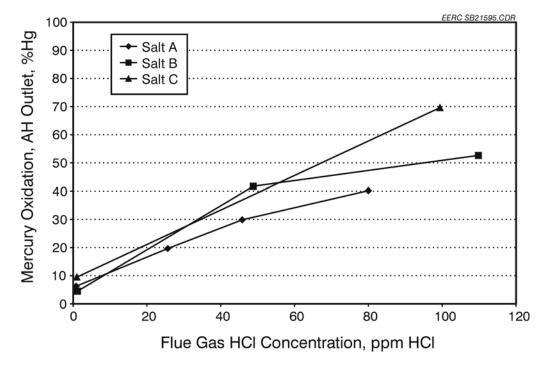


Figure 4. Comparison of mercury oxidation and HCl flue gas content for a range of salt injections at a North Dakota lignite-fired power plant (8).

collection area (SCA) of 375 ft 2 /1000 acfm. The wet FGD for SO $_2$ control utilizes alkaline ash and lime. The MRY Plant fires North Dakota lignite coal from the Kinneman Creek and Hagel seams at the Center Mine. This plant and configuration is ideal for testing Hg oxidation and Hg control in a wet scrubber. The high-temperature environment in the cyclone will easily vaporize and transform the chlorine species into highly reactive radical forms. The system has been tested for Hg speciation and control.

Recently, flue gas sampling for speciated Hg was conducted on MRY Unit 2 at the ESP inlet, FGD inlet, and stack from October 22 through November 14, 2002. The sampling was carried out using both the OH method and CMMs (9). A schematic of the plant and sample locations is provided in Figure 5. The sampling involved OH sampling at the ESP inlet, FGD inlet, and stack. In addition to OH sampling, two CMMs, one at the FGD inlet and one at the stack, were used to monitor speciated Hg levels. The CMMs were operated to obtain 20 days of data at the two locations.

The average Hg speciation results from Unit 2 OH flue gas sampling are summarized in Figure 6A. The average Hg emissions at the stack were 95% Hg 0 . Two CMMs were operated at the FGD inlet and stack locations of Unit 2 to gather Hg variability data. Statistical analysis of the CMM data indicates that the average Hg concentration was $10.7 \pm 2.7 \,\mu\text{g/m}^3$ (90th percentile) at the FGD inlet and $9.3 \pm 2.2 \,\mu\text{g/m}^3$ at the stack. Hg-level fluctuations due to minor coal changes as well as other variability in plant operations were found to fall within 24% of the average. A Hg balance for MRY Unit 2 (10) was determined by comparing the rate of Hg entering the plant to the rate of Hg leaving the plant. The resulting material balances ranged from 102% to 103%.

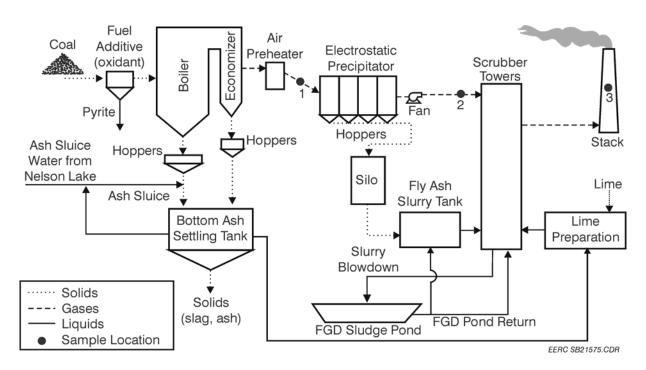


Figure 5. Schematic for MRY Unit 2 showing sampling locations.

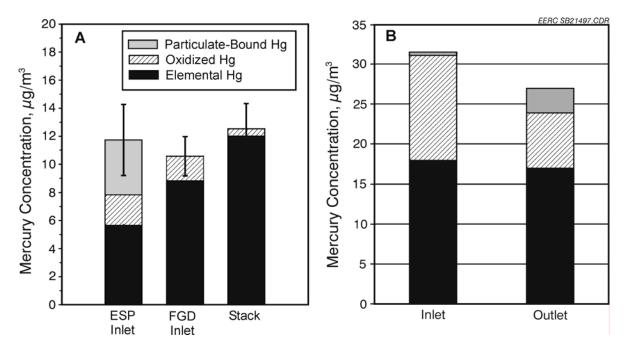


Figure 6. A) MRY OH mercury data obtained in October–November 2002 and B) OH mercury data for Monticello (ICR data).

The second site is the Monticello Unit 3 power plant located near Mt. Pleasant, Texas. This site is also well characterized for Hg speciation, emissions, and variability. In addition, it provides an opportunity to test the Hg oxidation technology on a Texas lignite. Figure 7 illustrates the Unit 3 gas path. Unit 3 has a 750-MW B&W wall-fired, Carolina-type universal pressure boiler that fires Texas lignite coal from the upper and lower Wilcox seam. The unit was placed in commercial operation in 1978 and fires 640 tons/hr of Texas lignite at full-rated load. Downstream of the air preheater, the gas flows through a cold-side ESP constructed by Hamon Research-Cottrell. The ESP has ten fields with an SCA of 900 ft²/1000 acfm. The ESP outlet temperature is nominally 300°F.

The results of Hg speciation measurements at the inlet and outlet of the scrubbers at the Monticello Unit 3 plant are shown in Figure 6B. The results of the OH method indicate that 57% of the total Hg is in the elemental form entering the wet FGD and that the Hg⁰ is not captured with the wet FGD. Results from the ICR tests at Monticello Unit 3 suggest that approximately 15% Hg removal across the FGD system, which is consistent with the trends for other units firing low-rank lignite coals.

3.0 EXPERIMENTAL

3.1 Objectives

The objective of this project is to demonstrate the effectiveness of chemical addition for reducing Hg emission from flue gas derived from lignite. Full-scale tests will be performed at

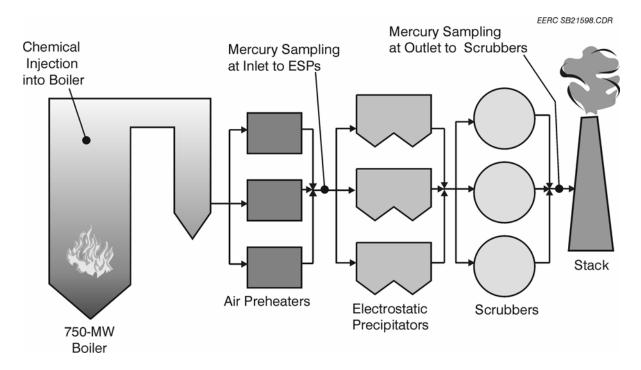


Figure 7. Plant schematic for Monticello Unit 3.

Minnkota Power Cooperative's MRY Unit 2 and TXU's Monticello Unit 3 to evaluate chemical addition performance across an ESP wet scrubber configuration.

The objective of the MRY Unit 2 testing is to determine the impact of chemical addition on Hg speciation, overall Hg removal from the flue gas using the combination of the ESP and wet scrubber, and the impact of the additive salts on corrosion and deposition on system components. The objective of the Monticello testing is to provide additional data on Hg oxidation and removal efficiency when a lignite coal from Texas is fired. Data from this program will be used to perform an economic analysis of the costs associated with full-scale implementation of a chemical addition system.

3.2 Planned Scope of Work

The scope of work is aimed at testing Hg oxidation technology for controlling Hg emissions at two lignite-fired power plants equipped with wet FGD systems. The plants include the MRY Unit 2 (cyclone-fired, North Dakota lignite, ESP, wet FGD) and Monticello Unit 3 (wall-fired, Texas lignite, ESP, wet FGD). The technology involves the injection of a chemical additive with the lignite or injection into the furnace to oxidize Hg upstream of a wet FGD system. The two plants with different firing systems and lignite types will be tested to determine the following: degree of mercury oxidation as a function of chemical addition rate, Hg removal efficiencies, economics, and BOP impacts. The additive will be added at rates equivalent to 300–1000 ppm chlorine in the coal during parametric testing, with a target of less than 500 ppm in the coal for the long term if selected. A second additive (SEA2) has repeatedly been shown to

be even more effective than chlorine and will therefore be tested as well. In addition, small amounts of solid oxidizing additive (SOA [activated carbon in this case]) will be added (<1 lb/Macf) to further enhance oxidation. The 2-month test will be conducted using the additive that performs the best during parametric testing. If fractions of lb/Macf of carbon are shown effective in enhancing the SEA impact, it will also be considered for the 2-month test.

4.0 RESULTS AND DISCUSSIONS

4.1 Summary of Activities Conducted October–December 2005

MRY

A site-specific test plan was developed that detailed the efforts to be conducted. A meeting on-site was conducted with MRY Plant personnel to discuss the detailed test plan. The test plan was made available to plant personnel and project sponsors for their review and comments. These comments were incorporated into a final version of the test plan. Detailed planning for the testing phase at the MRY Plant is well underway. The schedule for this testing is rigorous and, because of a scheduled unit outage, needs to be completed prior to the outage scheduled for June 9–16, 2005.

Corrosion probes were installed at the MRY Plant in the economizer, airheater inlet and airheater outlet November 18, 2004, and were removed January 20, 2005, for a total exposure time of 63 days under baseline conditions.

Installation of the corrosion probes with coupons exposed to long-term test conditions will occur in late May at the MRY Plant. Analysis of all corrosion probe coupons will be performed when the long-term testing is completed.

Monticello

Planning is progressing for testing at Monticello which will follow the MRY tests. The test plan for Monticello will be drafted next quarter.

4.2 Results from MRY Testing

No data were generated for this quarter.

4.3 Results from Monticello Testing

No data were generated for this quarter.

5.0 CONCLUSIONS/PLANNED ACTIVITIES FOR NEXT QUARTER

5.1 Task 1 – MRY Testing

- A 2-month test of chemical addition will be performed to enhance Hg oxidation and capture in wet FGD injection at the MRY Plant. During testing, the impact of chemical addition on Hg speciation and overall Hg removal from the flue gas using the combination of the ESP and wet scrubber will be measured. The additive will be added to the boiler with the coal feed. The testing will be conducted on run-of-mine (ROM) lignite. All efforts will be made to obtain samples of lignite during the course of testing.
- To accommodate any unforeseen delays and complete the work before the deadline, the tentative start of baseline testing has been set as the week of February 28, 2005, with injection systems and sampling instrumentation installed and tested the previous week.
- Field testing will be initiated in the next quarter.

5.2 Task 2 – Monticello Testing

• Full-scale tests will be performed at TXU's Monticello Station Unit 3 (Fall 2005) to evaluate the effectiveness of chemical addition on Hg control across an ESP wet scrubber configuration. Parametric tests will be used to determine the optimal process conditions for each material and will be used to establish the conditions for each extended test set to last 2 weeks.

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